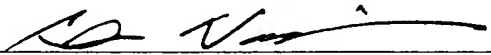


## Verified Translation

I, Shigemitsu Nakajima, hereby declare the following:

I am knowledgeable in Japanese and English. I have reviewed Certified Priority Document No. 2002-242216 filed on August 22, 2002 and believe the attached document to be an accurate translation thereof.

All statements made herein of my own knowledge are true and all statements made on information and belief are believed to be true. Further, these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Date: November 10, 2006 

Signature of Declarant

Patent Application 2002-242216(JP 242216/2002)

[Document Name] Patent application  
[Reference Number] P0001298  
[Addressee] Director General of the Patent Office  
[Inventor]  
[Address] c/o Sumitomo Mitsui Polyolefin Co.  
580-30, Nagaura, Sodegaura-shi, Chiba  
[Name] Yoshio SUGUMOTO  
[Inventor]  
[Address] c/o Sumitomo Mitsui Polyolefin Co.  
580-30, Nagaura, Sodegaura-shi, Chiba  
[Name] Ikunori SAKAI  
[Inventor]  
[Address] C/o Toyota Jidosha Kabushiki Kaisha  
1 Toyota-cho, Toyota-shi, Aichi  
[Name] Yusuke Umetani  
[Inventor]  
[Address] C/o Toyota Jidosha Kabushiki Kaisha  
1 Toyota-cho, Toyota-shi, Aichi  
[Name] Naoki Shoji  
[Inventor]  
[Address] 9-1, Tatuta Minami 6-chome, Ikaruga-cho,  
Ikoma-gun, Nara  
[Name] Keita Nagano  
[Inventor]  
[Address] Hakusuiryo, 3-30, Akebonogawa Higashi,  
Yao-shi, Osaka  
[Name] Takao Mizoshita  
[Inventor]  
[Address] C/o Dainichiseika Color & Chemicals Mfg. Co.,  
Ltd.,  
7-6, Nihonbashi Bakuro-cho 1-chome, Chuo-ku,  
Tokyo  
[Name] Isamu Yamaguchi

[Applicant]  
 [Identification No.] 000005887  
 [Name] MITSUI CHEMICALS, INC.

[Applicant]  
 [Identification No.] 000003207  
 [Name] TOYOTA JIDOSHA KABUSHIKI KAISHA

[Applicant]  
 [Identification No.] 399054321  
 [Name] TOYO ALUMINIUM KABUSHIKI KAISHA

[Applicant]  
 [Identification No.] 000002820  
 [Name] DAINICHISEIKA COLOR & CHEMICALS MFG.  
 CO., LTD.

[Agent]  
 [Identification No.] 100075524  
 [Patent Attorney]  
 [Name] Shigemitsu NAKAJIMA

[Agent Appointed]  
 [Identification No.] 100070493  
 [Patent Attorney]  
 [Name] Kano YAMAGUCHI

[Charge]  
 [Deposit Account] 059846  
 [Amount of Deposit] 21000 yen

[List of Filed Documents]  
 [Name of Document] Specification 1  
 [Name of Document] Abstract 1

[Number of General Power of Attorney] 9807060

[Proof] Necessary

[Name of Document] SPECIFICATION

[Title of the Invention]

RESIN COMPOSITION FOR AUTOMOTIVE PARTS

[Scope of Claims]

5 [Claim 1]

A resin composition for automotive parts comprising a polypropylene resin composition and an aluminum flake pigment, wherein

(1) the above polypropylene resin composition comprises 50 to 70% by weight of either of crystalline propylene/ethylene block copolymer (a-1) or blend of (a-1) and a crystalline propylene homopolymer (a-2) having not greater than an equivalent weight of (a-1), 18 to 25% by weight of elastomeric polymer (b), and 15 to 25% by weight of inorganic filler (c),

(wherein the total amount of the respective components is made 100% by weight) ; and

(2) the above aluminum flake pigment comprises aluminum flakes whose surface is coated with a polymer containing as constituent units acrylic acid, an acrylic acid ester, epoxylated polybutadiene and divinylbenzene.

10 [Claim 2]

The resin composition for automotive parts according to claim 1, wherein 0.005 to 10 parts by weight of the above aluminum flake pigment is contained based on 100 parts by weight of the polypropylene resin composition.

[Claim 3]

The resin composition for automotive parts according to either of claim 1 or 2, wherein the above polypropylene

resin composition has a melt flow rate (ASTM D-1238, 230°C, 2160g load) of 30 to 70 g/10 min, a flexural modulus (ASTM D-790) of 1900 to 3000 MPa, and a brittleness temperature (ASTM D-746) of -10 to -40°C.

[Claim 4]

The resin composition for automotive parts according to any one of claim 1 to 3, wherein the above crystalline propylene/ethylene block copolymer (a-1) is composed of a propylene homopolymer portion and a propylene/ethylene random copolymer portion, and has the melt flow rate (ASTM D-1238, 230°C, 2160 g load) of 70 to 130 g/10 min, and wherein the isotactic pentad fraction (mmmm fraction) in the propylene homopolymer portion, as measured by <sup>13</sup>C-NMR, is not less than 97%, and the content of the propylene/ethylene random copolymer portion is 5 to 20% by weight.

[Claim 5]

The resin composition for automotive parts according to any one of claim 1 to 4, wherein the above crystalline propylene homopolymer (a-2) has an isotactic pentad fraction (mmmm fraction) of not less than 97%, and a melt flow rate (ASTM D-1238, 230°C, 2160 g load) of 100 to 300 g/10 min.

[Claim 6]

The resin composition for automotive parts according to any one of claims 1 to 5, wherein the above elastomeric polymer (b) comprises a copolymer rubber (b-1) of ethylene and an  $\alpha$ -olefin of at least 6 carbon atoms having a melt flow rate (ASTM D-1238, 230°C, 2160 g load) of 0.5 to 10 g/10 min, an ethylene/ $\alpha$ -olefin/nonconjugated polyene random copolymer (b-2) having a melt flow rate (ASTM D-1238,

230°C, 2160 g load) of not greater than 1 g/10 min, and a hydrogenated block copolymer (b-3) that is a hydrogenated product of a block copolymer containing a polymer block of a monovinyl-substituted aromatic hydrocarbon compound and a polymer block of a conjugated diene compound.

[Claim 7]

The resin composition for automotive parts according to claim 6, wherein the above elastomeric polymer (b) contains, in 100% by weight of the polypropylene resin composition: 10 to 20% by weight of the copolymer rubber (b-1) of ethylene and an  $\alpha$ -olefin having not less than 6 carbon atoms; 1 to 5% by weight of the ethylene/ $\alpha$ -olefin/nonconjugated polyene random copolymer(b-2) ; and, 1 to 10% by weight of the hydrogenated block copolymer(b-3).

[Claim 8]

The resin composition for automotive parts according to either of claim 6 or 7, wherein the above hydrogenated block copolymer (b-3) is a styrene/ethylene/butene/styrene block copolymer, a styrene/ethylene/propylene/styrene block copolymer or a styrene/ethylene/propylene block copolymer.

[Claim 9]

The resin composition for automotive parts according to any one of claims 1 to 8, wherein the above inorganic filler (c) is talc.

[Claim 10]

The resin composition for automotive parts according to any one of claims 1 to 9, wherein the above aluminum flake pigment contains 0.5 to 20 parts by weight of a polymer

containing as constituent units an acrylic acid, an acrylic acid ester, an epoxylated polybutadiene and divinylbenzene, based on 100 parts by weight of aluminum flakes.

[Detailed explanation of the invention]

[0001]

[Technical field of the invention]

The present invention relates to resin composition for automotive parts comprising polypropylene resin mainly and relates more in detail to a resin composition suitable for producing automotive parts having a good metallic appearance.

[0002]

10 [Technical background of the invention]

Polypropylene resins are utilized as molding materials in various fields, including convenience goods, kitchenware, wrapping films, automotive parts, machine parts, electrical parts, etc. Practically, propylene resins are used in the form of compositions by adding there various compounding ingredients or additives, depending on performances required for individual products. In the fields of, for example, automotive exterior parts, etc. that require mechanical strength, polypropylene resin compositions wherein elastomers, talc, etc. are formulated have been utilized.

20 [0003]

In view of recent trends in large-sized, thin-wall automotive exterior parts and a simplified process for manufacturing parts, it is required for the polypropylene resins to have more improved mechanical and physical properties such as impact resistance, flexural modulus,

etc., to hardly cause flow marks or weld marks and to have high flowability enabling to cope with paintless finish.

[0004]

On the other hand, unity in design with the body part  
5 weighed heavily in automotive exterior parts. Such unity was achieved so far, either by blending polypropylene resins with various colorants and then molding the blend, or by painting molded articles in the same color as the body part.

Taking into account the simplified manufacturing process  
10 and recyclability of materials, such a polypropylene resin composition that can deal with non-painting and shows an good visual appearance, has now been demanded.

[0005]

In order to meet the demand for non-painting, there was  
15 proposed , for example, a technique of blending polypropylene resins with aluminum flakes frequently applied to paints for body, thereby imparting a metallic appearance similar to metallic coatings. However, because of poor compatibility with polypropylene resins, aluminum flakes  
20 used for paints tend to separate from polypropylene resins and readily agglomerate during the processing of melt molding, which makes difficult to stably produce molded articles exhibiting a good metallic appearance. Especially in polypropylene resins having improved flowability to deal  
25 with large-sized or thin-wall automotive exterior parts, a tendency to cause separation and agglomeration of aluminum flakes is observed notably.

[0006]

[The problem of the invention to be solved]



It is an object of the present invention to provide a resin composition for automotive parts, which has excellent mechanical strength, physical properties and flowability, shows improved dispersibility of an aluminum flake pigment and enables to produce molded products showing a good metallic appearance.

[0007]

[Means to solve the problem]

That is, the present invention relates to a resin composition for automotive parts which comprises a composition comprising a polypropylene resin composition and an aluminum flake pigment, wherein

(1) the above polypropylene resin composition comprises 50 to 70% by weight of crystalline propylene/ethylene block copolymer (a-1) or a blend of (a-1) and a crystalline propylene homopolymer (a-2) of not greater than an equivalent weight of (a-1), 18 to 25% by weight of elastomeric polymer (b) and 15 to 25% by weight of inorganic filler (c) (wherein the total amount of the individual components is made 100% by weight), and

(2) the above aluminum flake pigment comprises aluminum flakes whose surface is coated with a polymer containing as constituent units an acrylic acid, an acrylic acid ester, epoxylated polybutadiene and divinylbenzene.

[0008]

It is desirable that 0.005 to 10 parts by weight of the above aluminum flake pigment are contained based on 100 parts by weight of polypropylene resin composition. Moreover it is desirable that the aluminum flake pigment contains 0.5

to 20 parts by weight of polymer containing as constituent units an acrylic acid, an acrylic acid ester, an epoxylated polybutadiene and divinylbenzene based on 100 parts by weight of the aluminum flake pigment

5 [0009]

It is preferable that the above polypropylene resin composition has 30 to 70 g/10 min of the melt flow rate (ASTM D-1238, 230°C, 2160 g load), 1900 to 3000 MPa of flexural modulus (ASTM D-790) and -10 to -40°C of  
10 brittleness temperature (ASTM D-746).

[0010]

It is preferable that the crystalline propylene/ethylene block copolymer (a-1) comprises a propylene homopolymer portion and a propylene/ethylene random copolymer portion,  
15 and has the melt flow rate (ASTM D-1238, 230°C, 2160 g load) of 70 to 130 g/10 min, the isotactic pentad fraction (mmmm fraction) in the propylene homopolymer portion of not less than 97% as measured by <sup>13</sup>C-NMR and the content of propylene/ethylene random copolymer portion of 5 to 20% by  
20 weight.

[0011]

It is preferable for the above crystalline propylene homopolymer (a-2) that the isotactic pentad fraction (mmmm fraction) is not less than 97% and the melt flow rate (ASTM  
25 D-1238, 230°C, 2160 g load) is 100 to 300 g/10 min.

[0012]

It is preferable that the above elastomeric polymer (b) comprises a copolymer rubber (b-1) of ethylene and an  $\alpha$ -olefin of at least 6 carbon atoms having a melt flow rate

(ASTM D-1238, 230°C, 2160 g load) of 0.5 to 10 g/10 min , an ethylene/ $\alpha$ -olefin/nonconjugated polyene random copolymer (b-2) having a melt flow rate (ASTM D-1238, 230°C, 2160 g load) of not greater than 1 g/10 min, and a hydrogenated  
5 block copolymer (b-3) that is a hydrogenated product of a block copolymer containing a polymer block of monovinyl-substituted aromatic hydrocarbon compound and a polymer block of conjugated diene compound. And It is also desirable that 10 to 20% by weight of (b-1), 1 to 5% by weight of (b-  
10 2) and 1 to 10% by weight of (b-3) are contained in 100% by weight of the polypropylene resin composition.

[0013]

As the hydrogenated block copolymer (b-3), styrene/ethylene/butene/styrene block copolymer,  
15 styrene/ethylene/propylene/styrene block copolymer or styrene/ethylene/propylene block copolymer are preferable and talc is preferable as an inorganic filler (c).

[0014]

[Specific explanation of the invention]

20 Next, the resin composition for automotive parts in accordance with the present invention and the individual components that constitute the resin composition are specifically described below.

[0015]

#### 25 Polypropylene resin composition

The polypropylene resin composition which can be used in the present invention comprises the following components in the ratios given below:

(1) 50 to 70% by weight, preferably 55 to 65% by weight of

the crystalline propylene/ethylene block copolymer (a-1) or a blend of (a-1) and the crystalline propylene homopolymer (a-2) of not greater than the equivalent weight of (a-1); (2) 18 to 25% by weight, preferably 19 to 24% by weight of the elastomeric polymer (b); and (3) 15 to 25% by weight, preferably 18 to 23% by weight of the inorganic filler (c), wherein the total amount of the individual components is 100% by weight.

10 [0016]

The polypropylene resin composition having the composition as described above is excellent in flowability upon molding, provides superior physical properties such as flexural modulus, impact resistance, hardness, brittleness temperature, etc., and gives a good balance among these physical properties. Therefore, the polypropylene resin composition can be advantageously used as a raw resin for injection molding, exhibits good moldability in injection molding and can easily manufacture injection molded products having excellent dimensional stability.

20 [0017]

In the polypropylene resin composition described above, the composition which satisfies the following physical properties is preferred as raw components that constitute the resin composition for automotive parts:

- (1) the melt flow rate (MFR: ASTM D-1238, 230°C, 2160 g load) is preferably 30 to 70 g/10 min, more preferably 35 to 60 g/10 min;
- (2) the flexural modulus (ASTM D-790) is preferably 1900 to

3000 MPa, more preferably 1900 to 2500 MPa; and,  
(3) the brittleness temperature (ASTM D-746) is preferably -  
10 to -40°C, more preferably -20 to -35°C.

[0018]

5 The crystalline propylene/ethylene block copolymer (a-1)  
is used either alone or as an admixture of the block  
copolymer (a-1) and the crystalline propylene homopolymer  
(a-2) of not greater than the equivalent weight of (a-1).

[0019]

10 The propylene/ethylene block copolymer (a-1) is composed  
of the propylene homopolymer portion and the  
propylene/ethylene random copolymer portion. The content of  
the propylene homopolymer portion is preferably 80 to 95% by  
weight, more preferably 87 to 92% by weight, and the content  
15 of the propylene/ethylene random copolymer portion is  
preferably 5 to 20% by weight, more preferably 8 to 13% by  
weigh. Herein, the total amount of the two components  
becomes 100% by weight.

[0020]

20 The contents of the two components described above can be  
measured by fractionating a sample of the block copolymer at  
room temperature using p-xylene as a solvent and determined  
from the results of fractionation. In one example of such  
measurement methods, 5 g of the block copolymer sample is  
25 completely dissolved in boiled p-xylene; after that, the  
temperature is lowered to 20°C, the resulting solution is  
allowed to stand overnight and then filtered to remove the  
insoluble matters. Subsequently, 1500 ml of methanol is  
added to the filtrate and the mixture is stirred. The

soluble matter is separated as precipitates, which are taken out by filtration and dried to give the p-xylene-soluble matter. Since the soluble matter is the propylene/ethylene random copolymer portion, the content of the  
5 propylene/ethylene random copolymer portion can be determined by weighing the portion.

[0021]

The propylene homopolymer portion preferably shows the isotactic pentad fraction (mmmm) as measured by  $^{13}\text{C}$ -NMR of  
10 preferably not less than 97%, more preferably not less than 97.5%. Herein, the isotactic pentad fraction (mmmm fraction) indicates a ratio of isotactic chains in a pentad unit in the crystalline polypropylene molecule chain, which is measured using  $^{13}\text{C}$ -NMR. Specifically, the isotactic  
15 pentad fraction is determined to be a ratio of the absorption peak, in a  $^{13}\text{C}$ -NMR spectrum, of the propylene monomer unit located at the center of consecutive five propylene monomer units, which are bonded to each other with meso form, to all absorption peaks in the methyl carbon  
20 region. It is desirable for the propylene homopolymer portion to have MFR (230°C, 2160 g load) of preferably 100 to 300 g/10 min, more preferably 120 to 250 g/10 min

[0022]

It is desirable that the propylene/ethylene random  
25 copolymer portion has an intrinsic viscosity  $[\eta]$  of preferably 6 to 9 dl/g, as measured at 135°C in decahydronaphthalene, and the ethylene content of preferably 20 to 40% by weight, more preferably 24 to 32% by weight.

[0023]

In the block copolymer (a-1), the content of ethylene units is preferably in the range of 1 to 10% by weight, more preferably 3 to 8% by weight. The content of ethylene units in the block copolymer can be determined by subjecting a pressed film of the block copolymer (a-1) sample to IR absorption spectrometry, that is, by measuring absorbance at  $1155\text{ cm}^{-1}$  based on the methyl group and absorbance based on the methylene group and applying Gardner's calibration curve thereto (I. J. Gardner et al., Rubber Chem. And Tech., 44, 1015, 1971).

[0024]

Further, as the crystalline propylene/ethylene block copolymer (a-1), there are employed those having a melt flow rate (MFR:  $230^{\circ}\text{C}$ , 2160g load) of preferably 70 to 130 g/10 min, more preferably 80 to 120 g/10 min, as measured in accordance with ASTM D-1238. The use of a block copolymer having MFR smaller than the range described above is not preferred, because such a resin composition for automotive parts becomes easier to cause flow marks or weld marks on the surface of finally obtained molded products, and a thermal shrinkage rate of the molded products tends to become large. The crystalline propylene/ethylene block copolymer (a-1) can be used singly or in combination of two or more.

[0025]

In the present invention, the blend wherein a half or less of the crystalline propylene/ethylene block copolymer (a-1) is replaced by the crystalline propylene homopolymer (a-2) having an isotactic pentad fraction of preferably not less

than 97%, more preferably not less than 97.5%, and a melt flow rate (MFR: 230°C, 2160 g load) of preferably 100 to 300 g/10 min, more preferably 120 to 250 g/10 min can be used instead of the crystalline propylene/ethylene block

5 copolymer (a-1).

[0026]

The crystalline propylene/ethylene block copolymer (a-1) can be produced by various methods. For example, the crystalline propylene/ethylene block copolymer (a-1) can be  
10 produced using conventionally known stereoregular catalysts for olefins, such as a Ziegler-Natta type catalyst, a metallocene type catalyst, etc. As an example of the methods for producing the block copolymer (a-1) using the Ziegler-Natta type catalyst, there can be cited the method  
15 for polymerizing propylene in the presence of a catalyst composed of, e.g., a solid titanium catalyst component, an organo-metallic compound catalyst component and, as required, an electron donor, followed by copolymerizing propylene and ethylene. The crystalline propylene  
20 homopolymer (a-2), which can be used together with the crystalline propylene/ethylene block copolymer (a-1), may be produced as well, using these stereoregular catalysts for olefins.

[0027]

25 The elastomeric polymer (b) is not particularly restricted in its kind and amount used. A preferred embodiment is a combination of the following polymers. That is, a preferred elastomeric polymer contains, in 100% by weight of the propylene polymer composition:



(1) preferably 10 to 20% by weight, more preferably 13 to 19% by weight of the copolymer rubber (b-1) of ethylene and an  $\alpha$ -olefin having not less than 6 carbon atoms;

(2) preferably 1 to 5% by weight, more preferably 2 to 5% by weight of the ethylene/ $\alpha$ -olefin/nonconjugated polyene random copolymer (b-2); and

(3) preferably 1 to 10% by weight, more preferably 3 to 8% by weight of a hydrogenated block copolymer(b-3) that is a hydrogenated product of a block copolymer containing the polymer block of monovinyl-substituted aromatic hydrocarbon compound and the polymer block of conjugated diene compound.

The elastomeric polymer may be used singly or in combination of two or more.

[0028]

The copolymer rubber (b-1) of ethylene and an  $\alpha$ -olefin having not less than 6 carbon atoms is a rubber-like copolymer obtained by copolymerization of ethylene with an  $\alpha$ -olefin having not less than 6 carbon atoms, preferably 6 to 12 carbon atoms, e.g., 1-hexene, 4-methyl-1-pentene, 1-octene, 1-decene, 1-dodecene, etc. It is desirable that the weight ratio of ethylene to the  $\alpha$ -olefin having not less than 6 carbon atoms (ethylene/ $\alpha$ -olefin having not less than 6 carbon atoms) in the copolymer is preferably in the range of 90/10 to 50/50, more preferably in the range of 80/20 to 60/40.

[0029]

As the copolymer rubber (b-1), the copolymer rubber having MFR (ASTM D-1238, 230°C, 2160 g load) of preferably 0.5 to 10 g/10 min, more preferably 1 to 8 g/10 min is used. When

the copolymer rubber having MFR within the range given above is used, rigidity or low temperature impact resistance of molded products finally obtained can be improved.

[0030]

5 Such an ethylene/ $\alpha$ -olefin copolymer rubber (b-1) can be prepared by copolymerization of ethylene and an  $\alpha$ -olefin having not less than 6 carbon atoms using a stereoregular catalyst for olefins. In particular, the ethylene/ $\alpha$ -olefin copolymer prepared using a single site catalyst has a narrow  
10 range of molecular weight distribution and compositional distribution, therefore it provides an excellent effect in improving impact resistance at low temperatures. Examples of such a single site catalyst include a metallocene type catalyst containing a metallocene compound wherein a  
15 compound having the cyclopentadienyl skeleton is coordinated on a transition metal such as zirconium metal, etc., and an organoaluminum oxy-compound, or the like.

[0031]

The ethylene/ $\alpha$ -olefin/nonconjugated polyene random  
20 copolymer (b-2) is a random terpolymer rubber of ethylene, an  $\alpha$ -olefin and a nonconjugated polyene. As the  $\alpha$ -olefin, there can be cited an  $\alpha$ -olefin having generally 3 to 20 carbon atoms, preferably 3 to 10 carbon atoms, and specific examples include propylene, 1-butene, 1-pentene, 1-hexene,  
25 1-octene, 1-decene, 4-methyl-1-pentene, etc.

[0032]

Examples of the above nonconjugated polyene include a cyclic nonconjugated diene such as 5-ethylidene-2-norbornene, 5-propylidene-2-norbornene, dicyclopentadiene,

5-vinyl-2-norbornene, 5-methylene-2-norbornene, 5-isopropylidene-2-norbornene, norbornadiene, etc.; a linear nonconjugated diene such as 1,4-hexadiene, 4-methyl-1,4-hexadiene, 5-methyl-1,4-hexadiene, 5-methyl-1,5-heptadiene, 6-methyl-1,5-heptadiene, 6-methyl-1,7-octadiene, 7-methyl-1,6-octadiene, etc.; a triene such as 2,3-diisopropylidene-5-norbornene, etc. Among these polyenes, 1,4-hexadiene, dicyclopentadiene and 5-ethylidene-2-norbornene are preferably used.

10 [0033]

As the ethylene/ $\alpha$ -olefin/nonconjugated polyene random copolymer (b-2), it is desirable to use the random copolymer showing MFR (ASTM D-1238, 230°C, 2160 g load) of preferably not more than 1 g/10 min, more preferably 0.1 to 0.5 g/10 min. When the random copolymer of MFR within the range given above is used, occurrence of flow marks or weld marks on the surface of molded products produced from the resin compositions finally obtained can be avoided.

[0034]

20 It is desirable for the copolymer (b-2) to have the copolymerization rate of ethylene to the  $\alpha$ -olefin preferably in the range of 90/10 to 40/60, more preferably 85/15 to 50/50, as expressed in a molar ratio (ethylene/ $\alpha$ -olefin). Moreover, a ratio of the nonconjugated polyene is preferably in the range of 1 to 40, more preferably in the range of 2 to 35, as expressed by an iodine value of the random copolymer (b-2).

25 [0035]

The representative examples of the ethylene/ $\alpha$ -olefin /

nonconjugated polyene random copolymer (b-2) include ethylene/propylene/diene terpolymer (EPDM) and ethylene/1-butene/diene terpolymer.

[0036]

5 The hydrogenated block copolymer (b-3) that is a hydrogenated product of block copolymer containing the polymer block of monovinyl-substituted aromatic hydrocarbon compound and the polymer block of conjugated diene compound is a hydrogenated product obtained by hydrogenation of the  
10 block copolymer represented by formula (1) or (2) below, at the Y portion thereof:



15

wherein X is the polymer block of monovinyl-substituted aromatic hydrocarbon, Y is the polymer block of conjugated diene, and n is an integer of 1 to 5, preferably 1 or 2.

[0037]

20 Examples of the monovinyl-substituted aromatic hydrocarbon, which constitutes the polymer block shown by X in the formula (1) or (2) above, include styrene or derivatives thereof, such as styrene,  $\alpha$ -methylstyrene, p-methylstyrene, chlorostyrene, a lower alkyl-substituted  
25 styrene, vinyl-naphthalene, etc. These monovinyl-substituted aromatic hydrocarbons may be used singly or in combination of two or more. Styrene is particularly preferable.

[0038]

Examples of the conjugated diene, which constitutes the

polymer block represented by Y in the formula (1) or (2) above, include butadiene, isoprene, chloroprene, etc. These dienes may be used singly or in combination or two or more. Particularly preferable is butadiene or isoprene. When  
5 butadiene is used as the conjugated diene, it is desirable that a ratio of the 1,2-bond in the polybutadiene block is preferably 20 to 80% by weight, more preferably 30 to 60% by weight.

[0039]

10 In the hydrogenated block copolymer (b-3), a hydrogenation degree in the conjugated diene polymer block (Y portion) is preferably not less than 90 mol%, more preferably not less than 95 mol%, the content of X portion is preferably 10 to 25% by weight, and MFR (ASTM D-1238, 190°C, 2160g load) is  
15 preferably not more than 15 g/10 min, more preferably 1 to 10 g/10 min. When the block copolymer having the content of X portion within the range given above is used, a thermal shrinkage rate of molded articles produced from the resin composition finally obtained becomes small and their  
20 brittleness temperature is low.

[0040]

Specific examples of the hydrogenated block copolymer (b-3) are styrene-based block copolymers such as styrene/ethylene/butene/styrene block copolymer (SEBS)  
25 obtained by hydrogenation of styrene/butadiene/styrene tri-block copolymer, styrene/ethylene/propylene/styrene block copolymer (SEPS) obtained by hydrogenation of styrene/isoprene/styrene tri-block copolymer, styrene/ethylene/propylene block copolymer (SEP) obtained by

hydrogenation of styrene/isoprene di-block copolymer, etc.  
[0041]

The block copolymer prior to the hydrogenation can be prepared, e.g., by conducting block copolymerization of the  
5 respective monomers in an inert solvent in the presence of a lithium catalyst or a Ziegler catalyst. Such a method is described in detail in, e.g., Japanese Patent Publication No. SHO 40-23798, etc.

The hydrogenation of the conjugated diene polymer block  
10 can be carried out by hydrogenating the block copolymer described above in an inert solvent in the presence of a known catalyst for hydrogenation. Such a method is described in detail in, e.g., Japanese Patent Publication Nos. SHO 42-8704, SHO 43-6636, SHO 46-20814, etc.

15 [0042]

The above hydrogenated block copolymer (b-3) is commercially available under trade names of Kraton G1657 (manufactured by Shell Chemical Co., Ltd., trademark), Septon 2004 (manufactured by Kuraray Co., Ltd., trademark),  
20 Tuftec H1052 and Tuftec 1062 (Asahi Kasei Corporation, trademark), etc., and these commercial products may be used.

[0043]

Examples of the inorganic filler (c) include talc, clay, calcium carbonate, mica, silicates, carbonates, glass  
25 fibers, etc. Among them, talc is particularly preferable. Talc having an average particle size of 1 to 10  $\mu\text{m}$ , preferably 2 to 6  $\mu\text{m}$ , as measured by laser analysis, is preferable as talc. The inorganic filler may be used singly or in combination of two or more.

[0044]

Aluminum flake pigment

The aluminum flake pigment which can be used in the present invention comprises aluminum flakes, whose surface  
5 is coated with a polymer containing as constituent units acrylic acid, an acrylic acid ester, epoxylated polybutadiene and divinylbenzene.

[0045]

Herein, aluminum flakes can be produced by methods  
10 conventionally known, for example, by milling or grinding atomized powders, aluminum foils, deposited aluminum foils, etc., using a ball mill, an attriter, a stamp mill, etc. Particularly preferable are aluminum flakes obtained by grinding aluminum powders prepared by atomization technique  
15 using a ball mill. The purity of aluminum is not particularly restricted, but aluminum may be in the form of alloy with other metals, so long as it is extendable. Examples of alloy metals include Si, Fe, Cu, Mn, Mg, Zn, etc.

20 [0046]

Examples of the acrylic acid ester include an alkyl acrylate, a hydroxyalkyl acrylate, an ester of acrylic acid and polyol, etc. As the epoxylated polybutadiene, epoxylated 1,2-polybutadiene having a polymerization degree  
25 of 3 to 10 is preferable.

[0047]

By incorporating acrylic acid and acrylic acid ester units into the polymer with which the surface of aluminum flakes is coated, a coat having transparency, adhesiveness,

weatherproof and chemical resistance is formed and corrosion resistance of the coat is improved by a cross-linking effect of the divinylbenzene unit contained. Furthermore, in the polymerization between the individual components, the  
5 reaction efficiency is enhanced by the polymerizable double bond that the epoxylated polybutadiene has.

[0048]

The amount of the polymer which coats the surface of aluminum flakes is preferably 0.5 to 20 parts by weight, more preferably 1 to 15 parts by weight, based on 100 parts  
10 by weight of aluminum flakes. Within the range above, a coat in a uniform thickness having the physical properties described above is formed on the surface of aluminum flakes to provide good and uniform compatibility with the  
15 polypropylene resin composition.

[0049]

An example of the methods for producing such surface-coated aluminum flakes is explained below. While aluminum flakes are suspended in an organic solvent which dissolves  
20 the respective component monomers but does not dissolve the formed polymer, the respective monomers of acrylic acid, acrylic acid ester, epoxylated polybutadiene and divinylbenzene are polymerized in the presence of a polymerization initiator, whereby the surface of aluminum  
25 flakes is coated with the polymer.

[0050]

Examples of the organic solvent which is usable in the reaction include an aliphatic hydrocarbon such as hexane, heptane, octane, cyclohexane, etc.; an aromatic hydrocarbon



such as benzene, toluene, xylene, etc.; a solvent mixture of an aliphatic hydrocarbon and an aromatic hydrocarbon, such as mineral spirit, etc.; a halogenated hydrocarbon such as chlorobenzene, trichlorobenzene, perchloroethylene, trichloroethylene, etc.; an alcohol such as methanol, ethanol, n-propanol, n-butanol, etc.; a ketone such as 2-propanone, 2-butanone, etc.; an ester such as ethyl acetate, propyl acetate, etc.; an ether such as tetrahydrofuran, diethyl ether, ethylpropyl ether, etc.

10 [0051]

It is desirable that the above organic solvent is used in an amount of 300 to 3000 parts by weight, preferably 500 to 1500 parts by weight, based on 100 parts by weight of aluminum flakes. When the organic solvent is used within the range above, a viscosity of the solvent is kept optimum so that reactants are uniformly dispersed and polymerization proceeds at an optimum reaction rate.

[0052]

Examples of the polymerization initiator include radical generators such as di-t-butyl peroxide, acetyl peroxide, benzoyl peroxide, lauroyl peroxide, cumyl hydroperoxide, t-butylhydroperoxide,  $\alpha, \alpha'$ -azobisisobutyronitrile, etc. The polymerization initiator is used preferably in an amount of 0.1 to 1 part by weight, based on 100 parts by weight of aluminum flakes. With the amount within the range above, the polymerization proceeds at an optimum rate, aluminum flakes do not agglomerate, and a good color hue is maintained.

[0053]

In general, the polymerization is carried out in an inert gas such as argon gas, nitrogen gas, etc. The temperature is preferably in the range of 60 to 200°C. When the polymerization is carried out within this temperature range, the polymerization rate is kept optimum so that the polymer produced deposits uniformly on the surface of aluminum flakes and adheres thereto. An appropriate time period for the polymerization is 0.5 to 24 hours. After the polymerization is completed, the solvent is removed through a filtering device, etc. to give a wet paste. The paste can be used as it is, as one component of the resin composition. [0054]

It is preferable for the aluminum flake pigment coated with such a polymer to have a particle diameter of 10 to 100  $\mu\text{m}$ . The aluminum flake pigment provides a good and uniform compatibility with the polypropylene resin composition. Furthermore, the metallic appearance and brightness on the surface imparted by the pigment are hardly affected by the polymer coating, and continue over a long period of time because of corrosion resistance and weatherproof that the coat possesses.

[0055]

#### Resin composition

The resin composition for automotive parts in accordance with the present invention is a composition comprising the polypropylene resin composition and the aluminum flake pigment described above. The aluminum flake pigment is contained preferably in 0.005 to 10 parts by weight, more preferably 0.1 to 2 parts by weight, based on 100 parts by

weight of the polypropylene resin composition.

[0056]

When a mixing ratio of the polypropylene resin composition and the aluminum flakes is within the range described above, it becomes easy to blend the two components uniformly. Therefore, high mechanical strength and flowability of the polypropylene resin composition are exhibited, and the metallic appearance and brightness of aluminum flakes are also displayed. Accordingly, the surface appearance of molded articles produced from the composition is excellent.

[0057]

When the polypropylene resin composition are obtained in the form of pellets wherein the components (a-1), (a-2), (b) and (c) are previously blended, the resin composition can be prepared by melt-kneading together the polypropylene resin composition pellets, the aluminum flake pigment and additives added as required, using mixing devices such as a Banbury mixer, a monoaxial extruder, etc. When a biaxial extruder is employed for mixing, it is desirable to adopt such a method that the polypropylene resin composition pellets are fed through a main feeder and the aluminum flake pigment is fed through a side feeder, in order to protect the surface resin-coated layer of the aluminum flake pigment. Also when the individual components that constitute the polypropylene resin composition are blended with the aluminum flake pigment all at once in one step, it is desirable to feed the aluminum flake pigment through a side feeder. The aluminum flake pigment itself may be used for mixing, and in order to improve its uniform dispersion,

a master batch of the pigment together with polyethylene, polypropylene, etc. may be previously prepared, followed by blending the master batch with the other components.

[0058]

5 It is desirable that the resin composition of the present invention is used in combination with colorants such as carbon black, titanium oxide, etc. If necessary and desired, other additives such as a heat resistant stabilizer, an antistatic agent, a weatherproof stabilizer, 10 a light stabilizer, an anti-aging agent, an antioxidant, a UV absorbent, a softener, a dispersant, a lubricant, etc., may further be combined in the resin composition within such a range that does not damage the objects of the invention.

[0059]

15 These resin compositions can be advantageously used for molding automotive parts, especially automotive exterior trims, e.g., for molding parts such as bumpers, overfenders, side moldings, rocker moldings, etc.

[0060]

## 20 Examples

Next, the present invention is described in more detail, by way of EXAMPLES but is not limited thereto.

[0061]

First, various raw materials used in EXAMPLES are 25 explained below.

### (1) Polypropylene resin composition

The crystalline propylene/ethylene block copolymer, the elastomeric polymer and the inorganic filler described below were blended in proportions shown in Table 1 to give two

polypropylene resin compositions (A-1 and A-2). The melt flow rate, flexural modulus and brittleness temperature were measured and the obtained results are also shown in Table 1.

[0062]

5 a. Crystalline propylene/ethylene block copolymer:

- MFR (230°C, 2160 g load): 100 g/10 min
- Propylene homopolymer portion: 90% by weight
- Isotactic pentad fraction (mmmm fraction): 98%
- Propylene/ethylene random copolymer portion: 10% by
- 10 weight
- Intrinsic viscosity  $[\eta]$ : 7.5 dl/g
- (135°C, as measured in decahydronaphthalene solvent)
- Ethylene content: 26% by weight

[0063]

15 b. Elastomeric polymer:

b-1: Ethylene/1-octene random copolymer rubber

- Polymer prepared using a metallocene catalyst
- 1-Octene content: 27% by weight
- MFR (230°C, 2160 g load): 2 g/10 min

20 [0064]

b-2: Ethylene/propylene/5-ethylidene-2-norbornene  
terpolymer

- MFR (230°C, 2160 g load): 0.4 g/10 min
- Propylene content: 28% by weight
- 25 · Iodine value: 15

[0065]

b-3: Styrene/ethylene/butene/styrene block copolymer (SEBS)

- MFR (230°C, 2160 g load): 4.5 g/10 min
- Styrene content: 20% by weight

[0066]

c. Inorganic filler: talc

· Average particle size: 4  $\mu\text{m}$ 

[0067]

5

Table 1

	Polypropylene resin composition A-1	Polypropylene resin composition A-2
Constitution of PP resin composition (% by weight)		
· Block copolymer	59	50
· Elastomeric polymer		
· b-1	13	22
· b-2	3	3
· b-3	5	5
· Talc	20	20
Physical properties of PP resin composition		
· MFR (g/10 min)	40	35
· Flexural Modulus (MPa)	1980	1600
· Brittleness temperature ( $^{\circ}\text{C}$ )	-30	-38

[0068]

(2) Aluminum flake pigment

## B-1: Surface-coated aluminum flakes

In a four-necked flask of 2 liter volume, 0.3 g of acrylic acid, 3.7 g of trimethylolpropane acrylate, 1.5 g of divinylbenzene, 3.2 g of epoxylated 1,2-polybutadiene, 200 g of aluminum flakes (average particle size of 60  $\mu\text{m}$ , manufactured by Toyo Aluminium Kabushiki Kaisha, G type) and 1150 g of mineral spirit were charged. After nitrogen gas was introduced, the mixture was thoroughly mixed while stirring. The temperature in the reaction system was elevated to 80°C, and 1.1 g of  $\alpha,\alpha'$ -azobisisobutyronitrile was added to the mixture. The reaction was continued at 80°C for 2 hours. The resulting slurry was subjected to solid-liquid separation to give wet resin-coated aluminum flake pigment having a solid content of 85% by weight. The amount of the resin coated was 3.5 g based on 100 g of aluminum flakes.

[0069]

Next, 20 parts by weight of the surface-coated aluminum flakes, 77 parts by weight of linear low density polyethylene and 3 parts by weight of magnesium stearate (manufactured by NOF Corporation) as a dispersant were blended and melt-kneaded to prepare the master batch of aluminum flakes.

[0070]

## 25 B-2: Aluminum flakes without surface coat treatment

Average particle size of 60  $\mu\text{m}$ , manufactured by Toyo Aluminium Kabushiki Kaisha, G type

[0071]

(3) Titanium oxide

CR-50 Titanium, available from Ishihara Sangyo Kaisha, Ltd.

Average particle size: 0.25  $\mu\text{m}$

Product with aluminum-treated surface

5 [0072]

(4) Carbon black

Product from Mitsubishi Chemical Corporation

Trade name: Carbon Black #45

Average particle size of 24  $\mu\text{m}$

10 RCF furnace type

[0073]

Next, in order to evaluate the physical properties of the resin compositions obtained in EXAMPLES and COMPARATIVE EXAMPLES and molded articles produced therefrom, various tests were performed by the methods described below.

(1) Melt flow rate (MFR):

The test was performed in accordance with ASTM D-1238.  
Measurement conditions: 230°C under the load of 2160 g.

(2) Flexural modulus:

20 The test was performed in accordance with ASTM D-790.

(3) IZOD Impact strength:

The test was performed in accordance with ASTM D-256 under the condition of notched.

[0074]

25 (4) Spiral flow length:

Using a mold for resin flow length measurement having a spiral flow path of 3 mm thick, 10 mm wide and 2000 mm long, the resin composition was molded at 230°C of resin temperature. The flow length (mm) of the molded product



obtained was measured and its length was made a spiral flow length.

[0075]

(5) Test on agglomeration of aluminum flakes

5 Using a mold of 3 mm thick, 120 mm wide and 130 mm long, equipped with a side gate of 4 mm wide and 2 mm high at the center of the lateral side in the width direction, the resin composition was molded at 230°C continuously by 100 shots. The presence or absence of agglomeration of aluminum flakes, 10 which would take place on the surface of the square plate molded at the 100th shot around the side gate toward the flowing direction, was visually inspected.

o: No agglomeration of aluminum flakes was observed on the surface of the molded square plate.

15 x: Agglomeration of aluminum flakes was observed on the surface of the molded square plate.

[0076]

#### EXAMPLE 1

The respective components described in Table 2 except for 20 the aluminum flake pigment (B-1) were blended in the amounts given using a tumbler. The blend was thus fed through a first feeding port (main feeder) of a biaxial extruder and the aluminum flake pigment was fed through a second feeding port (side feeder). These components were melt-kneaded to 25 prepare the resin composition. MFR (230°C, 2160 g load) of the resin composition was 40 g/10 min.

Using this resin, various physical properties were measured and the appearance was evaluated. The results are shown in Table 2. The results of Table 2 reveal that the

resin composition exhibited high flowability and excellent mechanical strength and physical properties, without causing agglomeration of aluminum flakes.

[0077]

5 COMPARATIVE EXAMPLE 1

The same procedures as in EXAMPLE 1 were performed, except that aluminum flakes (B-2) without any surface treatment was used in place of the aluminum flake pigment (B-1) in EXAMPLE 1.

10 The aluminum agglomeration test was performed using the resin composition. Agglomeration of aluminum flakes occurred on the surface of the molded article produced from the resin composition.

[0078]

15 COMPARATIVE EXAMPLE 2

The same procedures as in EXAMPLE 1 were performed, except that the polypropylene resin composition (A-2) was used in place of polypropylene resin composition (A-1) in EXAMPLE 1.

In this resin composition, the flexural modulus decreased.

20

25

[0079]

Table 2

	EXAMPLE 1	COMPARATIVE EXAMPLE 1	COMPARATIVE EXAMPLE 2
Composition of resin (parts by weight)			
· Polypropylene resin composition			
(A-1)	100	100	-
(A-2)	-	-	100
· Aluminum flakes			
(B-1)	0.9	-	0.9
(B-2)	-	0.9	-
· Titanium oxide	0.5	0.5	0.5
· Carbon black	0.1	0.1	0.1
Physical property of composition			
· MFR (g/10 min)	40	40	35
· Spiral flow length (mm)	144	144	142
· Flexural modulus (MPa)	2050	2080	1620
· IZOD impact strength (J/m)	434	443	50
· Agglomeration of aluminum flakes	o	x	o

[0080]

[The effect of the invention]

According to the present invention, the resin composition  
5 blended uniformly in all over can be obtained by dispersing  
an aluminum flake in the polypropylene resin composition  
uniformly.

Therefore, the surface of the final molded articles  
produced from resin composition shows metallic appearance  
10 and brightness that then aluminum flake has, and shows a  
good appearance.

In addition, resin composition of the present invention  
has superior mechanical strength properties as shown in  
flexural modulus and izod impact strength. It also can  
15 avoid outbreak of flow mark and weld mark because it has a  
high fluidity as shown in spiral flow length. Therefore, the  
resin composition is suitable for production of automotive  
parts especially exterior parts of molded articles.

[Name of Document] ABSTRACT

[Abstract]

[Problem to be solved]

5     To provide a resin composition for automotive parts, which is excellent in mechanical strength, physical properties and flowability, and enables to produce molded products showing good metallic appearance.

[Means for solution]

10     It is a resin composition comprising polypropylene resin composition and an aluminum flake pigment. As polypropylene resin composition, a composition comprising a crystalline propylene/ethylene block copolymer, an elastomeric polymer and an inorganic filler is used. Further as an aluminum  
15 flake pigment, aluminum flakes whose surface is coated with a polymer containing as constituent units acrylic acid, an acrylic acid ester, epoxylated polybutadiene and divinylbenzene are used.

[Selected Drawing]

20     None.